



INVESTOR IN PEOPLE

**CERTIFIED COPY OF  
PRIORITY DOCUMENT**

101 002

09/769 561

The Patent Office  
Concept House  
Cardiff Road  
Newport  
South Wales  
NP10 8QQ

the undersigned, being an officer duly authorised in accordance with Section 74(1) and (4) of the Deregulation & Contracting Out Act 1994, to sign and issue certificates on behalf of the Comptroller-General, hereby certify that annexed hereto is a true copy of the documents as originally filed in connection with the patent application identified therein.

also certify that the attached copy of the request for grant of a Patent (Form 1/77) bears a correction, effected by this office, following a request by the applicant and agreed to by the Comptroller-General.

in accordance with the Patents (Companies Re-registration) Rules 1982, if a company named in this certificate and any accompanying documents has re-registered under the Companies Act 1985 with the same name as that with which it was registered immediately before re-registration save for the substitution as, or inclusion as, the last part of the name of the words "public limited company" or their equivalents in Welsh, references to the name of the company in this certificate and any accompanying documents shall be treated as references to the name by which it is so re-registered.

in accordance with the rules, the words "public limited company" may be replaced by p.l.c., p.l.c. or PLC.

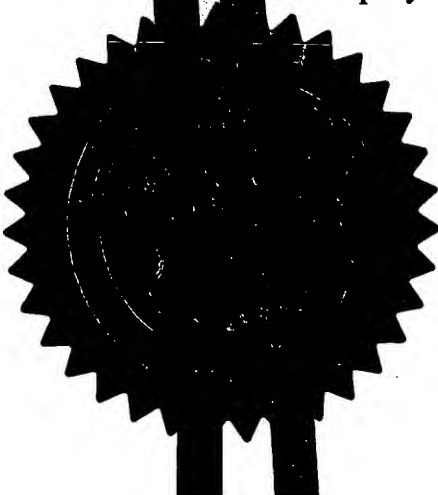
Re-registration under the Companies Act does not constitute a new legal entity but merely subjects the company to certain additional company law rules.

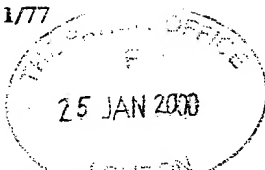
**BEST AVAILABLE COPY**

Signed

*Andrew Gusey*

Dated 26 May 2004





**Request for grant of a patent**

(See the notes on the back of this form. You can also get an explanatory leaflet from the Patent Office to help you fill in this form)

**The Patent Office**

Cardiff Road  
Newport  
Gwent NP9 1RH

- ### 1. Your reference

66/67/46271GB

2. Patent application number

(The Patent Office will fill in this part)

1. 2. 3. 4. 5. 6. 7. 8. 9. 10. 11. 12. 13. 14. 15. 16. 17. 18. 19. 20. 21. 22. 23. 24. 25. 26. 27. 28. 29. 30. 31. 32. 33. 34. 35. 36. 37. 38. 39. 40. 41. 42. 43. 44. 45. 46. 47. 48. 49. 50. 51. 52. 53. 54. 55. 56. 57. 58. 59. 60. 61. 62. 63. 64. 65. 66. 67. 68. 69. 70. 71. 72. 73. 74. 75. 76. 77. 78. 79. 80. 81. 82. 83. 84. 85. 86. 87. 88. 89. 90. 91. 92. 93. 94. 95. 96. 97. 98. 99. 100. 101. 102. 103. 104. 105. 106. 107. 108. 109. 110. 111. 112. 113. 114. 115. 116. 117. 118. 119. 120. 121. 122. 123. 124. 125. 126. 127. 128. 129. 130. 131. 132. 133. 134. 135. 136. 137. 138. 139. 140. 141. 142. 143. 144. 145. 146. 147. 148. 149. 150. 151. 152. 153. 154. 155. 156. 157. 158. 159. 160. 161. 162. 163. 164. 165. 166. 167. 168. 169. 170. 171. 172. 173. 174. 175. 176. 177. 178. 179. 180. 181. 182. 183. 184. 185. 186. 187. 188. 189. 190. 191. 192. 193. 194. 195. 196. 197. 198. 199. 200. 201. 202. 203. 204. 205. 206. 207. 208. 209. 210. 211. 212. 213. 214. 215. 216. 217. 218. 219. 220. 221. 222. 223. 224. 225. 226. 227. 228. 229. 230. 231. 232. 233. 234. 235. 236. 237. 238. 239. 240. 241. 242. 243. 244. 245. 246. 247. 248. 249. 250. 251. 252. 253. 254. 255. 256. 257. 258. 259. 260. 261. 262. 263. 264. 265. 266. 267. 268. 269. 270. 271. 272. 273. 274. 275. 276. 277. 278. 279. 280. 281. 282. 283. 284. 285. 286. 287. 288. 289. 290. 291. 292. 293. 294. 295. 296. 297. 298. 299. 300. 301. 302. 303. 304. 305. 306. 307. 308. 309. 310. 311. 312. 313. 314. 315. 316. 317. 318. 319. 320. 321. 322. 323. 324. 325. 326. 327. 328. 329. 330. 331. 332. 333. 334. 335. 336. 337. 338. 339. 340. 341. 342. 343. 344. 345. 346. 347. 348. 349. 350. 351. 352. 353. 354. 355. 356. 357. 358. 359. 360. 361. 362. 363. 364. 365. 366. 367. 368. 369. 370. 371. 372. 373. 374. 375. 376. 377. 378. 379. 380. 381. 382. 383. 384. 385. 386. 387. 388. 389. 390. 391. 392. 393. 394. 395. 396. 397. 398. 399. 400. 401. 402. 403. 404. 405. 406. 407. 408. 409. 410. 411. 412. 413. 414. 415. 416. 417. 418. 419. 420. 421. 422. 423. 424. 425. 426. 427. 428. 429. 430. 431. 432. 433. 434. 435. 436. 437. 438. 439. 440. 441. 442. 443. 444. 445. 446. 447. 448. 449. 450. 451. 452. 453. 454. 455. 456. 457. 458. 459. 460. 461. 462. 463. 464. 465. 466. 467. 468. 469. 470. 471. 472. 473. 474. 475. 476. 477. 478. 479. 480. 481. 482. 483. 484. 485. 486. 487. 488. 489. 490. 491. 492. 493. 494. 495. 496. 497. 498. 499. 500. 501. 502. 503. 504. 505. 506. 507. 508. 509. 510. 511. 512. 513. 514. 515. 516. 517. 518. 519. 520. 521. 522. 523. 524. 525. 526. 527. 528. 529. 530. 531. 532. 533. 534. 535. 536. 537. 538. 539. 540. 541. 542. 543. 544. 545. 546. 547. 548. 549. 550. 551. 552. 553. 554. 555. 556. 557. 558. 559. 560. 561. 562. 563. 564. 565. 566. 567. 568. 569. 570. 571. 572. 573. 574. 575. 576. 577. 578. 579. 580. 581. 582. 583. 584. 585. 586. 587. 588. 589. 590. 591. 592. 593. 594. 595. 596. 597. 598. 599. 600. 601. 602. 603. 604. 605. 606. 607. 608. 609. 610. 611. 612. 613. 614. 615. 616. 617. 618. 619. 620. 621. 622. 623. 624. 625. 626. 627. 628. 629. 630. 631. 632. 633. 634. 635. 636. 637. 638. 639. 640. 641. 642. 643. 644. 645. 646. 647. 648. 649. 650. 651. 652. 653. 654. 655. 656. 657. 658. 659. 660. 661. 662. 663. 664. 665. 666. 667. 668. 669. 670. 671. 672. 673. 674. 675. 676. 677. 678. 679. 680. 681. 682. 683. 684. 685. 686. 687. 688. 689. 690. 691. 692. 693. 694. 695. 696. 697. 698. 699. 700. 701. 702. 703. 704. 705. 706. 707. 708. 709. 710. 711. 712. 713. 714. 715. 716. 717. 718. 719. 720. 721. 722. 723. 724. 725. 726. 727. 728. 729. 730. 731. 732. 733. 734. 735. 736. 737. 738. 739. 740. 741. 742. 743. 744. 745. 746. 747. 748. 749. 750. 751. 752. 753. 754. 755. 756. 757. 758. 759. 760. 761. 762. 763. 764. 765. 766. 767. 768. 769. 770. 771. 772. 773. 774. 775. 776. 777. 778. 779. 780. 781. 782. 783. 784. 785. 786. 787. 788. 789. 790. 791. 792. 793. 794. 795. 796. 797. 798. 799. 800. 801. 802. 803. 804. 805. 806. 807. 808. 809. 810. 811. 812. 813. 814. 815. 816. 817. 818. 819. 820. 821. 822. 823. 824. 825. 826. 827. 828. 829. 830. 831. 832. 833. 834. 835. 836. 837. 838. 839. 840. 84

0001699.8

3. Full name, address and postcode of the or of each applicant (underline all surnames)

~~HEATRIC (UK) LTD~~  
~~46 HOLTON ROAD~~  
~~HOLTON HEATH~~  
~~POOLE~~  
~~DORSET BH16 6LT~~

Meggitt (UK) Limited  
Farms House, Cowgrove  
Wincborne, Dorset  
BH21 4EL

Patents ADP number (if you know it)

**If the applicant is a corporate body, give the country/state of its incorporation**

CF  $\frac{14}{31}$

04274866001  
FBI/DOJ

- #### 4. Title of the invention

### CATALYTIC BED REACTOR

5. Name of your agent (if you have one)

**"Address for service" in the United Kingdom  
to which all correspondence should be sent  
(including the postcode)**

FITZPATRICKS  
CARDINAL COURT  
23 THOMAS MORE STREET  
LONDON  
E1W 1XU

695009

Patents ADP number (if you know it)

6. If you are declaring priority from one or more earlier patent applications, give the country and the date of filing of the or of each of these earlier applications and (if you know it) the or each application number

Country

Priority application number  
(if you know it)

Date of filing  
(day / month / year)

7. If this application is divided or otherwise derived from an earlier UK application, give the number and the filing date of the earlier application

Number of earlier application

Date of filing  
(day / month / year)

8. Is a statement of inventorship and of right to grant of a patent required in support of this request? (Answer 'Yes' if:

YES

- a) any applicant named in part 3 is not an inventor, or  
b) there is an inventor who is not named as an applicant, or  
c) any named applicant is a corporate body.  
See note (d))

Patents Form 1/77

9. Enter the number of sheets for any of the following items you are filing with this form. Do not count copies of the same document:

Continuation sheets of this form

|             |     |
|-------------|-----|
| Description | 19  |
| Claim(s)    | 0   |
| Abstract    | 0   |
| Drawing(s)  | 515 |

10. If you are also filing any of the following, state how many against each item.

|  |   |
|--|---|
| Priority documents   | 0 |
| Translations of priority documents   | 0 |
| Statement of inventorship and right to grant of a patent (Patents Form 7/77) | 0 |
| Request for preliminary examination and search (Patents Form 9/77)           | 0 |
| Request for substantive examination (Patents Form 10/77)                     | 0 |
| Any other documents (please specify)   | 0 |

11. I/We request the grant of a patent on the basis of this application.

|           |  |             |
|-----------|--|-------------|
| Signature |  | Date        |
|           | FITZPATRICKS   | 25 JAN 2000 |

12. Name and daytime telephone number of person to contact in the United Kingdom
- CHRIS WAY  
0207 522 9001

**Warning**

After an application for a patent has been filed, the Comptroller of the Patent Office will consider whether publication or communication of the invention should be prohibited or restricted under Section 22 of the Patents Act 1977. You will be informed if it is necessary to prohibit or restrict your invention in this way. Furthermore, if you live in the United Kingdom, Section 23 of the Patents Act 1977 stops you from applying for a patent abroad without first getting written permission from the Patent Office unless an application has been filed at least 6 weeks beforehand in the United Kingdom for a patent for the same invention and either no direction prohibiting publication or communication has been given, or any such direction has been revoked.

**Notes**

- If you need help to fill in this form or you have any questions, please contact the Patent Office on 0645 500505.
- Write your answers in capital letters using black ink or you may type them.
- If there is not enough space for all the relevant details on any part of this form, please continue on a separate sheet of paper and write "see continuation sheet" in the relevant part(s). Any continuation sheet should be attached to this form.
- If you have answered 'Yes' Patents Form 7/77 will need to be filed.
- Once you have filled in the form you must remember to sign and date it.
- For details of the fee and ways to pay please contact the Patent Office.

## CATALYTIC BED REACTOR

### Field of the Invention

The present invention is applicable in the field of industrial chemistry and especially relates to improvements in chemical reactors. In general the invention relates to process control of temperature in a chemical reaction system and processing plant. Particularly the invention provides a reactor suitable for rapid conversion of a fluid reactant within the reactor in which the reactant temperature is maintained at a desired profile indirectly by means of a heat exchanging fluid.

### Background of the Invention

In the majority of chemical processes there is a heat demand or a need to dissipate heat. Therefore, a wide range of chemical plant is involved in containing or conveying fluids which must at some stage of the process be either heated or cooled. One might consider furnaces, evaporators, distillation units, dryers and reaction vessels as plant where heat transfer manifests itself as a design and operational problem. In particular many industrial chemical processes employ reactors in which reactions are effected under given temperature and pressure conditions in the presence of a catalyst. Almost all these reactions generate or absorb heat i.e. they are exothermic or endothermic. The cooling effects for endothermic reactions generally adversely affect the rate of reaction and the corresponding parameters such as conversion and selectivity of the products from the reaction. The uncontrolled heating of exothermic

reactions generally leads to damage to the associated apparatus as the temperature can rise to a very high level. The reaction in such a case may become uncontrolled (so-called "run away reaction") and lead to unwanted by-products and undesired effects, such as deactivation of a process catalyst. Furthermore, whilst an ideal catalyst does not theoretically participate in a reaction in reality many catalysts become degraded or poisoned as the reaction progresses and on an industrial scale the costs associated with catalyst re-generation or replacement represent a significant burden. It will be understood that such costs also must include the down time for the plant or restrictions on capacity if a particular reactor has to be off-line for catalyst re-generation purposes. Therefore, it is desirable to prolong the life of a catalyst bed in view of the significant cost benefits that may be obtained overall. The invention to be described hereinafter is ideally suited to use in catalytic reactor design but can be adapted for other purposes. No distinction is made as to the application thereof to batch or continuous reaction systems.

Those skilled in the art recognise that it is beneficial for the changes in temperature resulting from the heating or cooling effects of the reactions to be controlled. It is well known that maintaining the temperature of the reaction at a given constant level may result in significant advantages to the reaction, such as improved conversion and selectivity, prolonged life of the catalyst and associated apparatus, reduced levels of unwanted by-products etc. In some cases, a slight

varying of the theoretical temperature profile may be more beneficial.

In order to effectively control the temperature of reactions within an acceptable range, the chemical industry has devised several arrangements, those commonly used being discussed in standard references and texts e.g. one might consider the general teachings by Octave LEVENSPIEL in Chapter 19 of Chemical Reaction Engineering. The relative merits of each approach is also discussed therein.

Conventionally, the temperature inside reactors has been controlled by passing an auxiliary heat exchanging fluid through tubes or between plates, same forming a heat transfer conductive medium or thermal bridge whilst separating the reaction species from the auxiliary heat exchange fluid. Thus it will be understood that in such an indirect heat transfer system there is on the one hand a process path or zone and on the other an auxiliary fluid path or zone separated by the tube wall(s) or plate surface(s).

Considering this well known concept in relation to packed catalytic bed reactors, reactant fluid is passed through the catalyst bed and heat of reaction therein is controlled by contacting the catalytic bed reaction zone with such auxiliary fluid containing tubes or plates. However, particularly for highly exothermic reactions, such an approach has not been found to be ideal since the packed bed often develops heat gradients, e.g. the catalyst bed will be cooler at its areas of contact with the said tubes or plates and hotter within its depth remote from said tubes or plates, permitting formation of

hot spots or moving hot fronts leading to variations in reaction progress within the bed as a whole. Thus at such a hot spot the reaction may proceed faster and hence catalyst therein will be more rapidly degraded. This  
5 will be particularly significant in the case of large plants.

An example of an arrangement of a reactor designed to offer more control over the reactant temperature is the staged adiabatic packed bed reactor. This system  
10 uses an arrangement wherein a number of discrete, spaced apart zones of reaction are provided with means therebetween to control the temperature of the products leaving a first zone of reaction prior to entering the next reaction zone. No heat exchanging means is provided  
15 to control the temperature of the reaction in the zones of the reaction. Thus the reactant fluid entering the reactor at a desired temperature passes through a packed bed containing catalyst. Upon exiting this first stage, the reactant gas and any products will have a temperature  
20 higher or lower than that of the initial temperature depending upon the reaction thermal characteristics. A heat exchanger then heats or cools the reactant gas to a second desired temperature, which may or may not be equivalent to the temperature of the first, before  
25 passing to the next packed bed i.e. the second stage. This sequence is repeated until the desired conversion is obtained. Thus the temperature profile of the reaction will be stepped within an acceptable range of temperature, and will therefore not be truly isothermal  
30 which is theoretically preferred.

An alternative proposal for a process and apparatus for controlling reaction temperatures is disclosed in US patent No. 5,600,053. This arrangement uses corrugated heat exchange plates spaced apart with each plate defining a boundary of a heat exchange flow channel on one side of the plate and a boundary of a reaction flow channel on the other. In the arrangement, a heat exchange fluid passes in the first of the aforementioned channels and a reactant stream passes through the second, preferably with a catalyst being present. This arrangement is intended to eliminate or minimise the typical step-wise approach to the so-called isothermal condition objective.

However, the arrangement proposed in US-A-5,600,053 requires adjacent corrugated plates to be joined together. For this purpose, smooth edges are provided to facilitate the assembly of superposed multiple plates to form channels. The plates are joined, such as by welding, along these smooth edges and hence the integrity of the seal of the channels formed by the corrugations in adjacent plates is not ideal, particularly where a large pressure differential exists between the heat exchange flow channels and the reaction flow channels since this will tend to urge the adjacent plates apart. This arrangement will thus place unnecessary constraints on parameters of the reaction, namely the relationship between the pressure of the heat exchanging fluid and that of the reactant gas.

An earlier system is described in US-A-5,073,352 which proposes an apparatus for conducting a process of reforming gasolines, under low pressure and in the



presence of at least one catalyst, in which heat required for the reaction is provided by a heat carrying fluid such as natural gas.

The apparatus described therein comprises a number of discrete reaction cells being arranged vertically and being of substantially parallelepipedic configuration. The cells are laterally spaced apart, thus forming channels therebetween for flow of the heat carrying fluid. The reforming catalyst-containing chambers are respectively either isothermal or adiabatic and dimensioned such that height (H), width (W) and thickness (T) satisfy the conditions  $H > W > T$ , and H is at least twice the value of W, W lying in the range of 50 mm to 10,000 mm (0.05 - 10 metres) and T lies in the range of 2 mm to 2,000 mm (0.002 - 2 metres). Thus there remains the possibility of hot spots and less than satisfactory thermal control in such large catalytic reactor volumes.

It is known to the man skilled in the art that the heat transfer coefficient in a packed bed is mainly dependent upon the catalyst particle size and the reactant fluid velocity through the catalytic bed. Unfortunately, both these parameters are process requirements and hence cannot be changed in order to improve the heat transfer coefficient in the packed bed, and hence in the reaction cells described in US 5,073,352. Additionally, it is difficult to move catalyst between narrow gaps or tubes, imposing limits on the dimensions of gaps or tubes through which catalyst is designed to flow.

Thus reactors of the known types according to the existing art have many significant limitations imposed on

the heat transfer capability. Plate reactors offer some advantages over tubular reactors on the auxiliary medium side, but the end result is not significant since the overall heat transfer coefficient is generally governed by the process side as discussed above. Tubular reactors on the other hand offer advantages over plate reactors with regard to mechanical capability, due to increased resistance to differential pressure between the reactant fluid and the heat exchanging fluid.

10        Considering the foregoing matters, it is an object of this invention to provide improvements in chemical plant design and methods of operation thereof with a view to obviating or mitigating the drawbacks of the existing or previously proposed designs and methods.

15        Particularly, it is an aim of the present invention to provide an apparatus and a process for the control of reaction temperature within an acceptable range during operation of the chemical process by an indirect heat transfer method using a heat exchanging fluid.

20        Another object of the invention to be described more particularly hereinbelow is to provide an apparatus permitting control of the reactant temperature closely within a desired profile, more specifically, aiming to maintain the temperature at a substantially constant level i.e. to offer attainment of an isothermal reaction zone in so far as is practical on an industrial scale.

25        It is a further object of the invention to provide chemical plant which is improved over known plant equipment in terms of both cost and space efficiency  
30        considerations.

### Summary of the Invention

The invention addresses the problems observed in the prior art by adopting the approach of staged adiabatic reactors and improving the performance thereof by design of a catalytic reactor comprising a catalyst bed and heat  
5 exchange means of the plate type in operative contact with the catalyst bed, wherein the heat exchange means is formed from a plurality of superposed metal plates wherein fluid flow channels have been chemically milled  
10 e.g. by etching, according to a pre-determined pattern, said channel-bearing plates being aligned during superposition to define discrete heat exchange pathways for fluids and diffusion bonded together.

The invention further addresses the drawbacks of the  
15 known art by providing a process for conversion of a fluid reactant which process according to the invention uses a catalytic reactor comprising catalyst bed and heat exchange means of the plate type in operative contact with said bed and having discrete fluid pathways for heat  
20 exchange between fluids at differing temperatures whilst avoiding mixing of the fluids, the said process providing the appropriate fluid reactant species to be converted in a catalytic reaction zone in the catalyst bed within the reactor and at a predetermined stage of reaction  
25 introducing at least a portion of the fluid reactant species into a reactant fluid pathway within said heat exchange means, and also introducing an auxiliary fluid at a temperature differing from that of the fluid reactant species into another fluid pathway within said  
30 heat exchange means and juxtaposed to the first whereby the discrete nature of the respective pathways permits

indirect heat exchange between the fluid reactant species, said process being optionally repeated in successive stages.

5 The residence time of the fluid reactant species within the catalytic reaction zone before introduction of at least a portion of same into the heat exchange means will be predetermined according to predicted reaction conditions and will be monitored in the usual way by observations made by a process operative or automatic  
10 device sensing of reaction conditions including pressure and particularly temperature, such observations or sensing permitting adjustment of the process to advance or retard the progress of the reactant fluid as is considered necessary.

15 According to a modification of the process additional fluid reactant species may be introduced at subsequent catalyst bed stages. Thus although it is envisaged that the process can be operated in a series of stages in the manner of the known staged adiabatic  
20 reactor systems the proposed reactor design permits greater control over the process not only in terms of heat management but also in terms of chemical reaction control.

25 Thus according to one aspect of the present invention, there is provided an apparatus for controlling the temperature profile of a reactant fluid in the presence of a catalyst during an endothermic or exothermic chemical reaction, comprising a reactor having reactant fluid inlet means and reactant fluid outlet  
30 means; catalytic beds being provided therebetween, spaced apart by a heat exchanger; said heat exchanger comprising

heat exchanging fluid inlet means, heat exchanging fluid outlet means, a first channel or set of channels for passage of the heat exchanging fluid, and a second channel or set of channels in communication with the adjacent catalytic beds to allow passage of the reactant fluid from one catalytic bed to the next, said second channel or set of channels not being in communication with the reactant fluid.

Preferably a screen made of a fine mesh lines the walls of the catalytic bed, and said walls are ideally formed at least in part by plates of the said heat exchangers. The mesh acts to resist migration of catalyst into the reactant fluid-receiving channels of the heat exchanger down stream of the catalytic reaction zone.

The heat exchanger of choice is one formed from a plurality of plates superposed and diffusion bonded to form a stack of plates, wherein fluid channels are defined in said stack by virtue of a pre-treatment of said plates wherein each plate is selectively configured according to the desired pattern of channels by a chemical treatment to remove surface material e.g. by chemical etching, to a desired depth. Such a pre-treatment of the plates is conducted in a manner analogous to manufacture of printed circuit boards (PCBs) and for this reason the reactor design described herein can be described as a printed circuit reactor (PCR). The proposed reactor design offers an infinite variety of auxiliary heat exchanging fluid and reactant fluid pathways of very small dimensions which enables significantly enhanced process control.

The or each heat exchanger stack may be formed from a length or block of superposed plates by division e.g. by cutting into individual slices of a desired dimension which enables very slim designs of great strength.

5        Thus in one construction, a first such channel or set of channels is perpendicular to a second such channel or set of channels. In an alternative construction, the respective channels are parallel. Naturally one would generally arrange the construction such that juxtaposed  
10 channels contain respectively reactant fluid species on the one hand and auxiliary fluid on the other to achieve the desired heat transfer. In this way temperature control is achieved indirectly without mixing of the reactant fluids with the auxiliary fluid media.

15        The profile of the channels, i.e. cross-sectional profile perpendicular to the flow path, is generally not critical but curved shapes are conventionally used and are relatively easy to form by chemical milling but other profiles could be adopted if desired by use of an  
20 appropriate tool in combination with the chemical process.

In use of such a PCR reactor, the heat exchanging fluid may be caused to flow in a direction substantially perpendicular to the flow of the reactant stream.  
25 Alternatively, the directions of flow may be substantially parallel and either co or counter current according to the operators choice taking account of the reaction process to be controlled.

The invention is primarily intended for use with  
30 reactions using heterogeneous catalyst systems.

Depending on the particular reaction, optimisation thereof may be obtained by providing more than one catalyst, and in particular by providing different catalysts in separate catalytic beds.

5       The heat exchanging fluid may be a liquid or gas, as is understood by those in this art. Such fluids typically include molten salts, molten metals or hot water to provide liquid auxiliary media or may be hot gases, steam or superheated steam whereby heat may be  
10 indirectly added to a reaction system. In the converse case chilled liquids or gases may be used. Chemical engineers will readily consider the wide range of auxiliary working fluids available and be aware of both sensible heat and latent heat considerations in matching  
15 the auxiliary fluid needed to the process demands of the reaction for conversion of the fluid reactant species.

Each heat exchanger stack or each auxiliary fluid media channel, or set of channels, may contain a different auxiliary fluid to optimise the temperature  
20 profile of the reactant fluid within the reactor.

Operation of the PCR is preferably such that the temperature of the fluid is below 800°C and the pressure below 500 bars.

According to another aspect of the invention, there  
25 is provided additional means to make the aforementioned reactor specifically suitable for use as a moving bed reactor, namely catalyst inlet means, catalyst outlet means and means for feeding new or regenerated catalyst into the catalyst inlet means, and further means to  
30 remove catalyst from the catalyst outlet means.

Preferably catalyst is allowed to progress towards the catalyst outlet under the influence of gravity. The man skilled in the art will be aware of alternative manners in which moving bed reactors may operate and the scope of the invention should not be restricted to the particular method described herein.

In the case of such a moving bed reactor, the bed width is a multiple of the catalyst diameter preferably at least 3 times that diameter.

According to a still further aspect of the invention, there is provided a process for indirectly controlling the temperature profile of a reaction fluid in the presence of a catalyst during an endothermic or exothermic chemical reaction, comprising passing a reactant fluid from a reactant fluid inlet means in a reactor to a first catalytic bed before passing through a first channel or set of channels in a heat exchanger and subsequently passing to a second catalytic bed; passing a heat exchanging fluid from a heat exchanging inlet means to a heat exchanging outlet means through a second channel or set of channels in the heat exchanger; and exchanging heat between the heat exchanging fluid and the reactant fluid whilst passing through the heat exchanger; the products of the reaction leaving the last catalytic bed being passed to a reaction fluid outlet means.

In an alternative embodiment, the process for controlling the temperature profile of a reaction fluid in the presence of a catalyst during an endothermic or exothermic reaction further comprises passing catalyst through the catalytic bed, catalyst leaving the bed being replaced by new or regenerated catalyst.



Preferably the catalyst is a heterogeneous catalyst.

In a preferred embodiment, the heat exchanging fluid flows in a direction substantially perpendicular to the flow of the reactant stream. Alternatively, the  
5 directions of flow may be substantially parallel and either co or counter current.

The heat exchanging fluid may be liquid or gas, in accordance with the prior art. Such fluids include molten salts and boiling water. Preferably the  
10 temperature of the fluid is below 800°C and the pressure below 500 bars.

It will be recognised that the invention in its broadest aspect provides a staged reaction system containing successive chemical reaction zones and heat  
15 transfer zones, the latter containing plate heat transfer surfaces bearing micro-channels etched therein according to a pre-determined pattern, and offering not only the optimum indirect heat transfer strategy but also the ability to individually prepare fluid reactants thermally  
20 for the next reaction zone by selection of the inlet channel and its relationship to adjacent auxiliary fluid channels. The system can be specifically designed to handle substances of different volatilities.

#### Description of the Drawings

25 By way of example, the embodiments of the invention will be described with reference to the accompanying drawings:

Figure 1 is a sectional side view of a reactor in accordance with the present invention;

Figures 2A and 2B show the temperature profile for an endothermic reaction in an adiabatic reactor and in a reactor in accordance with the present invention, respectively; and

5        Figures 3A and 3B show the temperature profile for an exothermic reaction obtained through a tubular reactor cooled with an auxiliary medium and a reactor in accordance with the present invention, respectively.

10                    Mode for Carrying out the Invention

Referring to the drawings, Figure 1 shows a reactor 1 provided with a reactant fluid inlet 2 and a reactant fluid outlet 3 through which a reactant fluid to be processed is passed.

15        The reactor 1 comprises at least one catalytic bed 4 in communication with the reactant fluid inlet 2. The catalytic bed 4 is substantially vertical and the chamber walls defined therefor are preferably smooth. These smooth walls facilitate the movement of the catalyst  
20 through the chamber where the apparatus is configured as a moving bed reactor.

Provided adjacent to the catalytic bed reactor 4 are heat exchangers 5. The heat exchangers 5 have at least two sets of channels formed therein, a first set of  
25 channels provided for passage of the reactant fluid from a first catalytic bed 4 to a second, and a second set of channels through which the heat exchanging fluid flows.

An optional screen of a fine mesh suitably dimensioned according to the size of the catalyst

particles is provided in this embodiment to cover the ends of the reactant fluid channels adjacent to the catalytic beds to prevent migration of catalyst into said channels, especially the down stream channels with respect to the catalyst to reduce the risk of blockages inhibiting flow.

Flow performance can be optimised and blockages decreased inside the channels, if the channel walls are smooth and free of crevices. The channels are formed by joining individual plates together, said plates having channels formed in at least one surface, such as by a milling process. In order to ensure the crevice free design, it is thus important that any joining process used to join adjacent plates together does not interfere with the machined channels. This therefore limits the acceptability of use of conventional welding processes and exposes the weakness of prior art proposals such as US-A-5,600,053. However, diffusion bonding processes, wherein the plates are placed under pressure and heated close to the melting temperature of the plate metal thereby encouraging grain growth across the boundary, allows such a crevice-free design. This also enables the plates to be joined adjacent each of the channels, increasing the mechanical capabilities of the channels and allowing greater pressure differentials between the pressure of the reactant fluid and the heat exchanging fluid. This design of heat exchanger has been proven by the designers of the proposed PCR since 1985 when Heatric first introduced its compact printed circuit heat exchangers (PCHEs). The application of the diffusion bonding technique is now understood in the art.

The design of such heat exchangers also facilitates a greater rate of heat exchange in a given volume, reducing the space requirements for a given reaction. Thus incorporating them in a custom reactor design offers  
5 hitherto unforeseen advantages.

Depending on the performance requirements of the reactor 1, additional catalytic beds 4 may be provided, spaced apart by additional heat exchangers 5. The final catalytic bed 4 in the series is in communication with  
10 the reactant fluid outlet means 3 to enable the exit of the products of the reaction from the reactor 1.

In the alternative embodiment wherein the apparatus is adapted to be suitable for use as a moving bed reactor, catalyst outlet means (not shown) are provided  
15 adjacent the lower end of each of the catalytic beds 4, through which the catalyst is urged by means of gravity. The catalyst leaving the catalytic beds 4 may be fed to a regenerator and then passed through catalyst inlet means (not shown) provided adjacent the top of each of the  
20 catalytic beds 4. Alternatively, new catalyst material may be passed through the catalyst inlet means, consequent to exit of catalyst through the outlet means.

Thus the apparatus of the invention facilitates a series of adiabatic reactions, the temperature of the  
25 reactant stream being altered between successive reactions to maintain the reaction temperature within an acceptable range and thus conform to the desired temperature profile.

Figures 2A and 2B relate to endothermic reactions.  
30 Figure 2A shows the temperature profile for an adiabatic.

reactor wherein the temperature decreases at a decreasing rate due to the cooling effects of the reaction.

Figure 2B shows a similar reaction performed in accordance with the present invention. An adiabatic  
5 reaction takes place in the first catalytic bed, the size of said bed having been determined by the desired temperature of the reactant fluid leaving the first bed. The reactant fluid is then heated in the first exchanger  
10 for the process up to a maximum value. The sequence is repeated with additional catalytic beds and heat exchangers until the desired conversion is obtained, the lower and upper values for the temperature range having been selected to optimise the process, said range being  
15 as narrow as required.

Figures 3A and 3B relate to exothermic reactions wherein heat is generated by the reaction. Figure 3A shows the temperature profile of the reactant fluid obtained in a tubular reactor, cooled by an indirectly  
20 heat exchanging fluid, such as molten salts. Similarly, Figure 3B shows the temperature profile of the reactant fluid for an exothermic reaction in a reactor in accordance with the present invention. Again, the reactant fluid enters a first catalytic bed wherein an  
25 adiabatic reaction takes place, raising the temperature of the fluid to a maximum permitted value before cooling of the fluid in a first heat exchanger. The sequence is again repeated until the required conversion is obtained.

The number of heat exchangers is calculated in  
30 accordance with methods known to the man skilled in the

art in order to avoid hotspots and to allow the process to run at a higher load of feed per unit of flow rate.

For example, phthalic-anhydride may be obtained by oxidation of ortho-xylene in the presence of air. For such a reaction, conventional tubular reactors are hot spot limited, and as a consequence, the maximum amount of oxide in air is limited to about 80 g/Nm<sup>2</sup>. Performing the same reaction in a reactor in accordance with the present invention allows much higher rates of reaction and consequently an increased throughput.

1/5

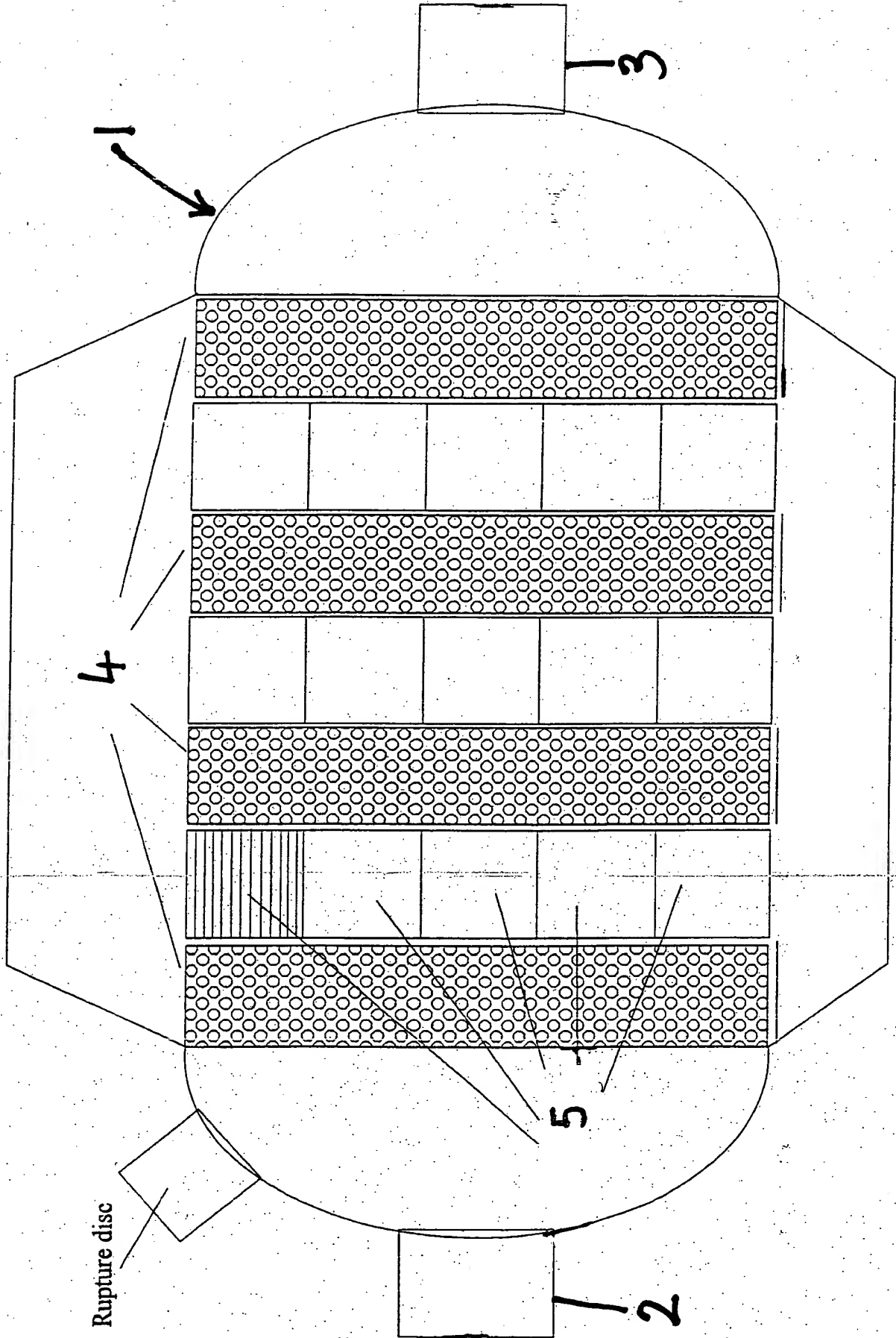


FIG 1

# Temperature profile in an adiabatic reactor

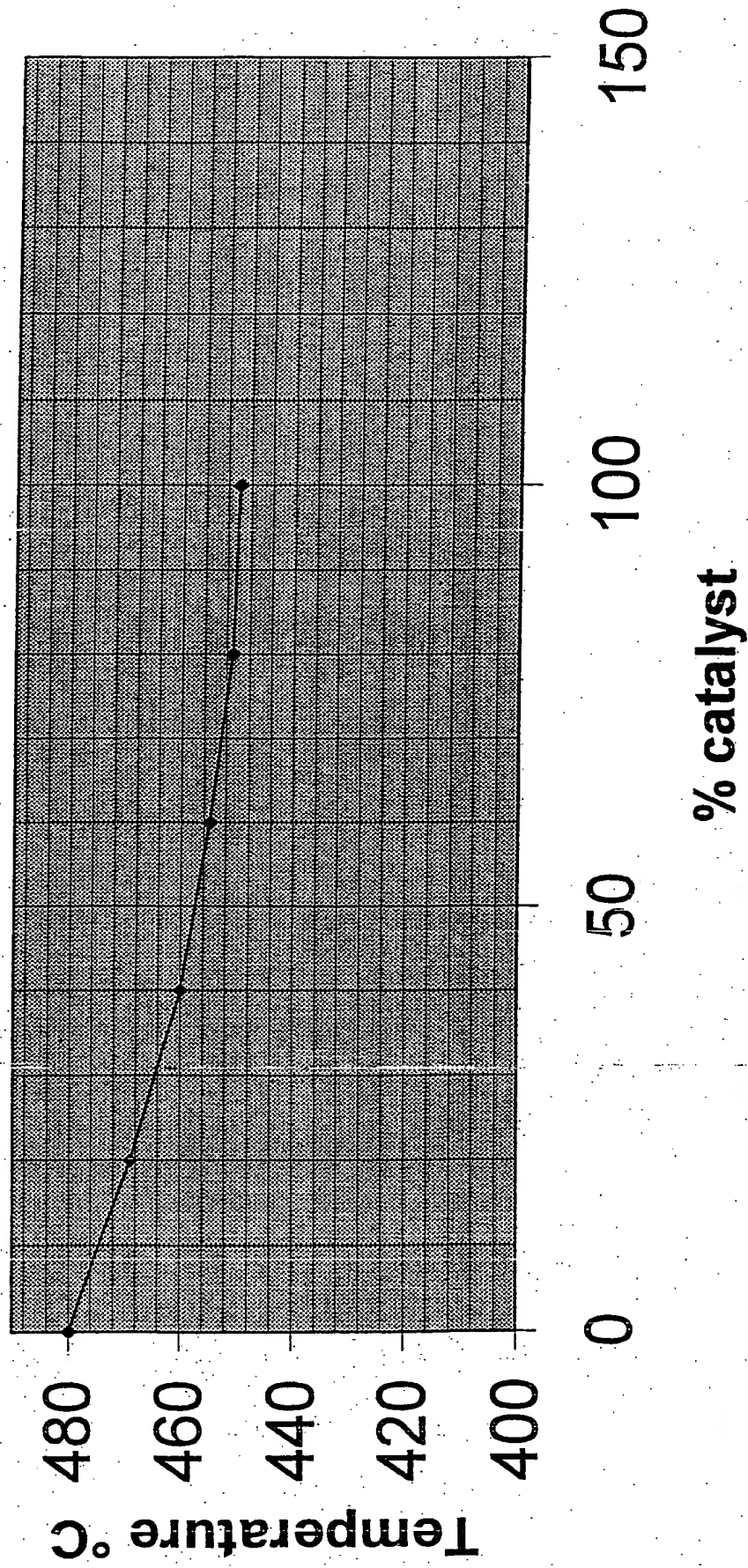


FIG 2.a



# Temperature profile in a PCHE reactor using 3 exchangers

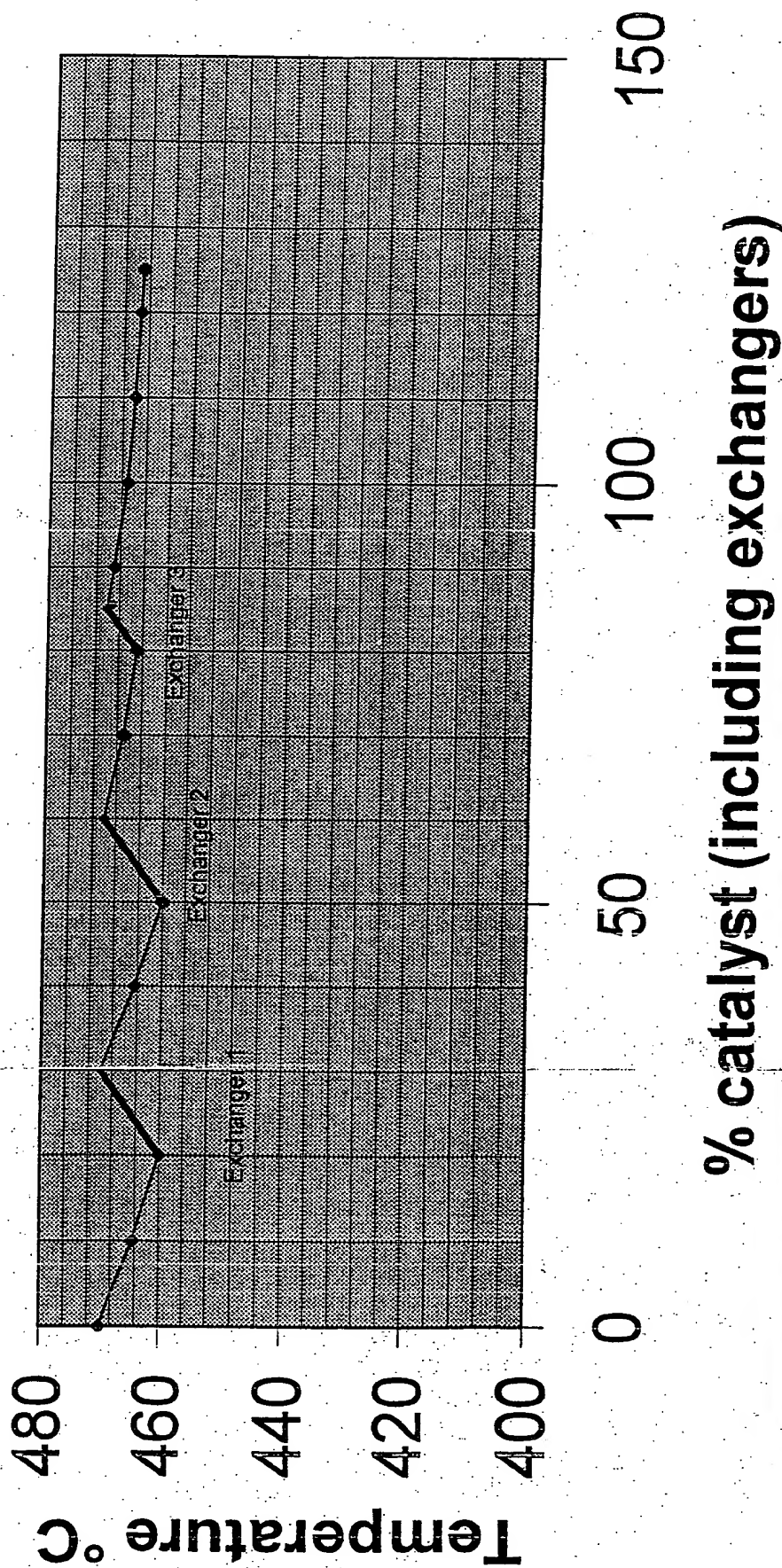


FIG 26.

# Temperature profile in a tubular reactor for an exothermic reaction

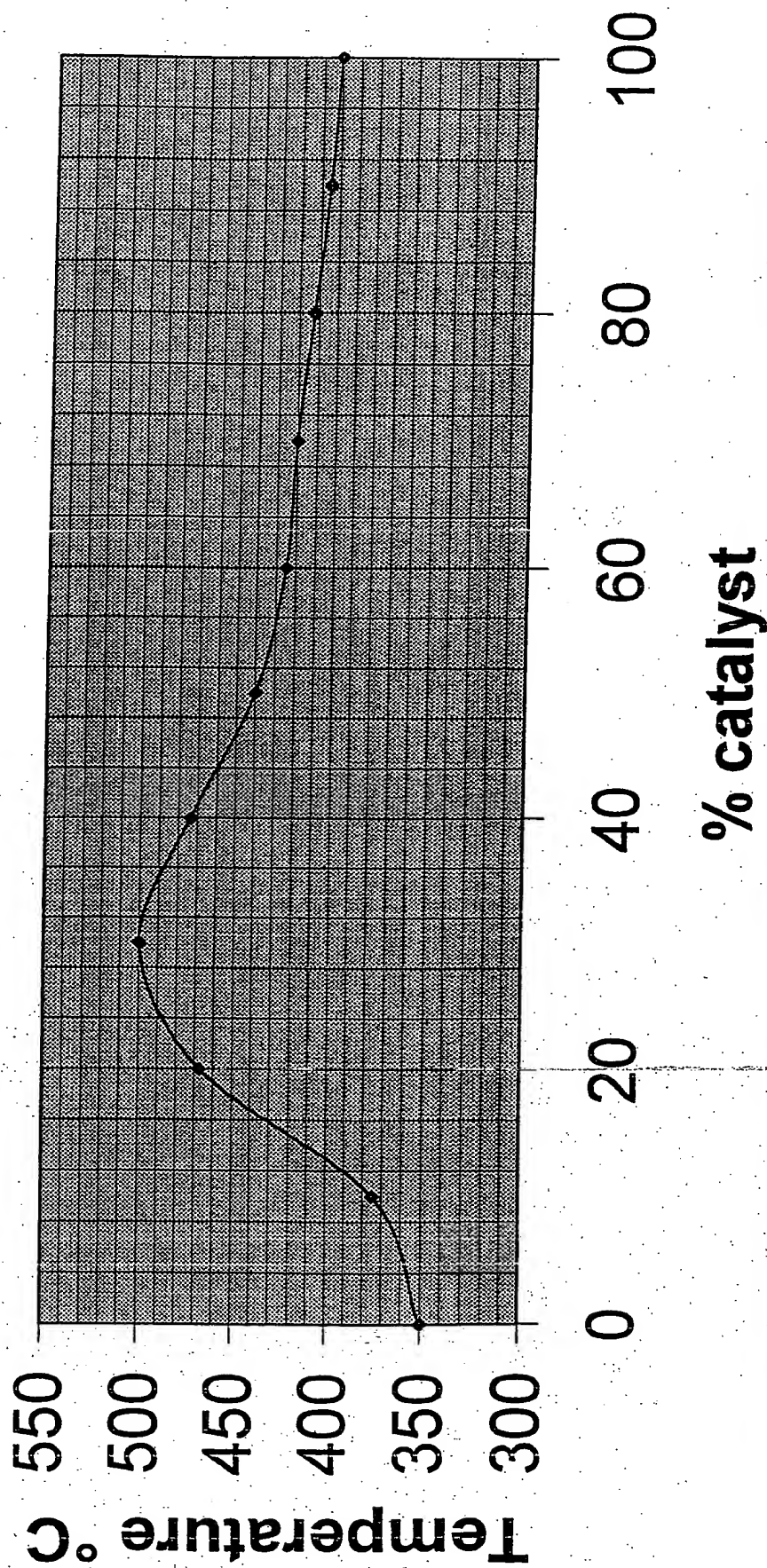


FIG 3a

# Temperature profile in a PCHE reactor for an exothermic reaction using 5 exchangers

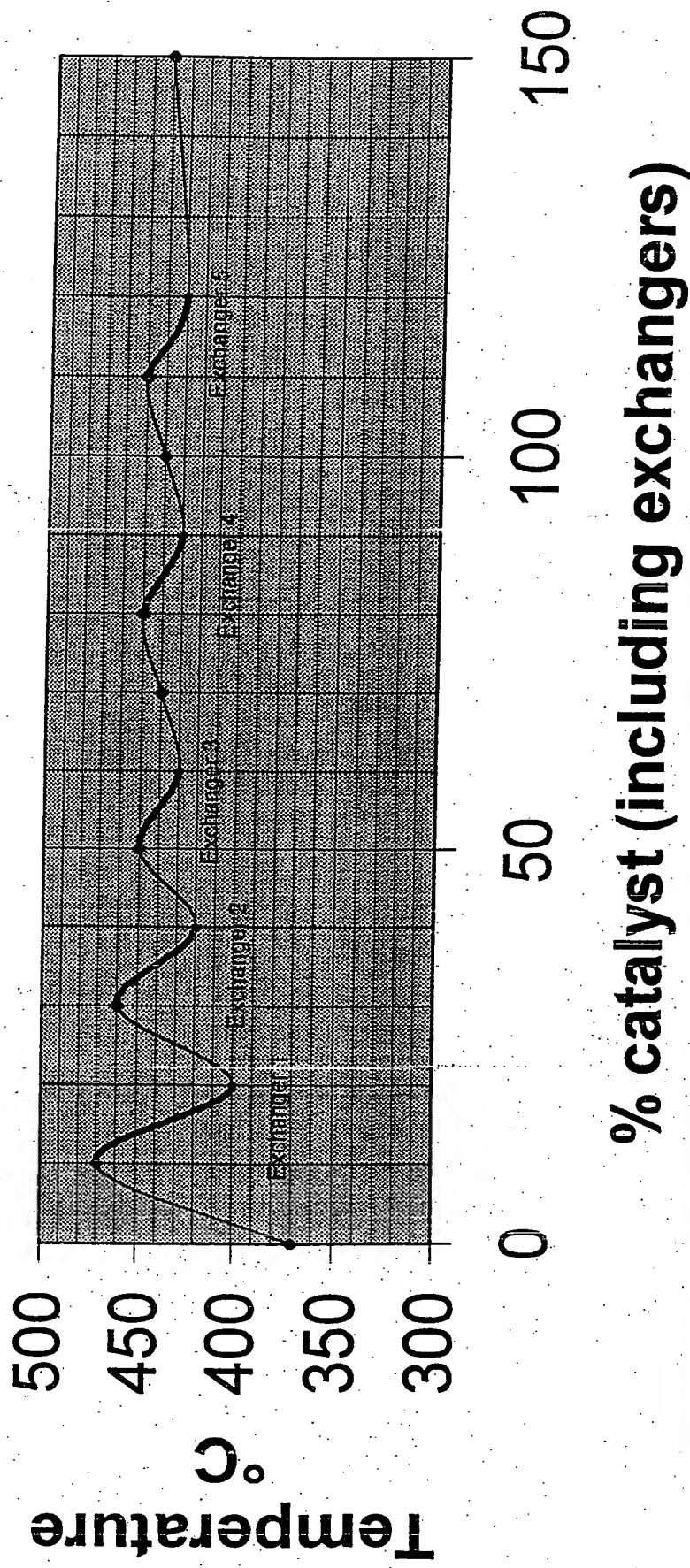


FIG 3.b

**This Page is Inserted by IFW Indexing and Scanning  
Operations and is not part of the Official Record**

**BEST AVAILABLE IMAGES**

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images include but are not limited to the items checked:

- ☐ BLACK BORDERS
- ☐ IMAGE CUT OFF AT TOP, BOTTOM OR SIDES
- ☒ FADED TEXT OR DRAWING
- ☐ BLURRED OR ILLEGIBLE TEXT OR DRAWING
- ☐ SKEWED/SLANTED IMAGES
- ☐ COLOR OR BLACK AND WHITE PHOTOGRAPHS
- ☐ GRAY SCALE DOCUMENTS
- ☒ LINES OR MARKS ON ORIGINAL DOCUMENT
- ☐ REFERENCE(S) OR EXHIBIT(S) SUBMITTED ARE POOR QUALITY
- ☐ OTHER: \_\_\_\_\_

**IMAGES ARE BEST AVAILABLE COPY.**

**As rescanning these documents will not correct the image problems checked, please do not report these problems to the IFW Image Problem Mailbox.**